

# Thermodynamic properties of ferromagnetic/superconductor/ferromagnetic nanostructures

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## Abstract

The theoretical description of the thermodynamic properties of ferromagnetic/superconductor/ferromagnetic (F/S/F) systems of nanoscopic scale is proposed. Their superconducting characteristics strongly depend on the mutual orientation of the ferromagnetic layers. In addition, depending on the transparency of S/F interfaces, the superconducting critical temperature can exhibit four different types of dependences on the thickness of the F-layer. The obtained results permit to give some practical recommendations for the spin-valve effect experimental observation. In this spin-valve sandwich, we also expect a spontaneous transition from parallel to anti-parallel ferromagnetic moment orientation, due to the gain in the superconducting condensation energy.

## I. INTRODUCTION

The peculiar character of the proximity effect in superconducting/ferromagnet (S/F) systems is due to the strong exchange field acting on the electrons in the ferromagnet and provoking the oscillatory-like behavior of the superconducting order parameter. Several in-

interesting phenomena inherent to the S/F hybrid structures have been predicted and subsequently observed on experiments: non-monotonous dependance of the critical temperature in S/F structures on the thickness of the ferromagnetic layer [1–7],  $\pi$ -junction realization in S/F/S systems [8–11] and local quasiparticle density of states oscillation in S/F structures [12–14].

In recent years, a great progress has been achieved in preparation of high quality hybrid S/F systems, especially high quality interfaces, which could be quite interesting for possible applications. In particular, a very promising system is the F/S/F spin-valve sandwich, where spin-orientation-dependant superconductivity has been predicted in [15–17].

In this article, we present the results of detailed theoretical studies of the properties of F/S/F systems containing a thin superconducting layer (compared to the superconducting coherence length). We analyze the influence of the F-layer thickness and the S/F interface transparency on the spin-valve superconductivity effect. The last part of the article is devoted to the thermodynamic properties of the spin-valve : we calculate the superconducting order parameter and the superconducting condensation energy for parallel and anti-parallel spin orientation of the F-layers. We discuss also the possibility of a spontaneous phase transition, by decreasing the temperature, from parallel to anti-parallel spin orientation.

## II. GENERAL EQUATIONS

We will concentrate on the studies of the properties of an F/S/F trilayer system with F-layers of thickness  $d_f$  and an S-layer of thickness  $d_s$ , see Fig. 1. Assuming that the dirty limit conditions are held in all layers, we may use the complete set of Usadel equations [18] in the superconducting layer and in the F-layers. In the superconducting layer the Usadel Green functions  $F$  and  $G$  satisfy

$$-\frac{D_s}{2}\overrightarrow{\nabla}\left[G(x,\omega)\overrightarrow{\nabla}F(x,\omega)-F(x,\omega)\overrightarrow{\nabla}G(x,\omega)\right]+\omega F(x,\omega)=\Delta(x)G(x), \quad (1)$$

in F-layers they verify [13]

$$(\omega + ih) F(x, \omega) - \frac{D_f}{2} \overrightarrow{\nabla} \left[ G(x, \omega) \overrightarrow{\nabla} F(x, \omega) - F(x, \omega) \overrightarrow{\nabla} G(x, \omega) \right] = 0, \quad (2)$$

and in both layers

$$G^2(x, \omega) + F(x, \omega) F^*(x, -\omega) = 1, \quad (3)$$

where  $D_s$  and  $D_f$  are the diffusion coefficients in S and F-layer respectively,  $\omega = 2\pi T(n + 1/2)$  are the Matsubara frequencies and  $h(x)$  is the exchange field in the F-layers. In the case of the parallel orientation of the magnetization of the F-layers, the exchange field is  $h(x) = h$  for  $x < -d_s/2$  and  $x > d_s/2$  whereas in the anti-parallel case  $h(x) = h$  for  $x > d_s/2$  and  $h(x) = -h$  for  $x < -d_s/2$ . The Usadel equations are completed by the self-consistency equation in the form [21]

$$\Delta \ln \frac{T}{T_c} + \pi T \sum_{\omega} \left( \frac{\Delta}{|\omega|} - F_s \right) = 0, \quad (4)$$

and by the boundary conditions at the S/F boundaries [19]

$$\begin{aligned} \frac{\partial F_s}{\partial x} &= \gamma \frac{\partial F_f}{\partial x}, \\ F_s &= F_f \pm \xi_f \gamma_B \frac{\partial F_f}{\partial x}, \end{aligned} \quad (5)$$

where  $\gamma = \frac{\sigma_s}{\sigma_f}$ ,  $\sigma_f$  ( $\sigma_s$ ) is the conductivity of the F-layer (S-layer above  $T_c$ ),  $\xi_f = \sqrt{\frac{D_f}{2h}}$ ,  $\xi_s = \sqrt{\frac{D_s}{2T_c}}$  is the superconducting coherence length of the S-layer, the parameter  $\gamma_B = \frac{R_b \sigma_f}{\xi_f}$ , where  $R_b$  is the S/F boundary resistance per unit area. In the second boundary condition, the sign before the spatial derivative of  $F_f$  depends on the relative orientation of the  $x$ -axis and the normal of the ferromagnet surface. If the normal is parallel to the  $x$ -axis ( $x = d_s/2$ ) the minus sign is required, in the other case ( $x = -d_s/2$ ) the positive sign is required. The parameter  $\gamma_B$  is directly related to the transparency of the interface  $T = \frac{1}{1+\gamma_B}$  [20]. The limit  $T = 0$  ( $\gamma_B = \infty$ ) corresponds to a vanishingly small boundary transparency, and the limit  $T = 1$  ( $\gamma_B = 0$ ) corresponds to a perfectly transparent interface. At the interface between the vacuum and the ferromagnet, the boundary condition is simply written as  $\frac{\partial F_f}{\partial x} = 0$ .

### III. USADEL EQUATIONS FOR THIN SUPERCONDUCTING INTERLAYER

The mutual influence of superconductivity and ferromagnetism reveals interesting effects for S-layer thickness smaller or of the order of magnitude of the superconducting coherence length  $\xi_s$ , otherwise, we have practically independent bulk superconductor and ferromagnetic systems. In addition, the case  $d_s \ll \xi_s$  has an analytical solution, that is the reason why we will suppose this condition to be satisfied in the following analysis. In this limit, the small spatial variations of the Green functions in S-layer can be taken into account by a simple expansion to the order  $x^2$

$$F_s = F_0 \left( 1 + \alpha x + \frac{\beta}{2} x^2 \right), \quad (6)$$

$$G_s = G_0 \left( 1 + ax + \frac{b}{2} x^2 \right), \quad (7)$$

where  $F_0$  and  $G_0$  are the values of the anomalous and normal Green functions at the center of the S-layer. Using (1) and (3), we finally obtain an effective Usadel equation for thin superconducting layers

$$\left[ \omega - \frac{D_s \beta}{2G_0} - \frac{D_s \alpha^2 F_0^2}{4G_0^3} \right] F_0 = \Delta G_0. \quad (8)$$

The coefficients  $\alpha$  and  $\beta$  in expression (8) have to be found by using the boundary conditions at the F/S interfaces. As it may be easily demonstrated from the boundary conditions, the term containing  $\alpha$  is by a factor  $(d_s/\xi_s) \ll 1$  smaller than the term with  $\beta$ , consequently this term can be neglected. Thus, in our approximation of thin S-layer, the Usadel equations take the following simple form

$$\left[ \omega - \frac{D_s \beta}{2G_0} \right] F_0 = \Delta G_0, \quad (9)$$

$$F_0^2 + G_0^2 = 1, \quad (10)$$

where the coefficient  $\beta$  plays the role of a pair-breaking parameter. The boundary conditions on the function  $F_s$ , following from (6), are

$$(F'_s/F_s)_{-d_s/2} = \alpha - d_s\beta/2, \quad (11)$$

$$(F'_s/F_s)_{d_s/2} = \alpha + d_s\beta/2.$$

By adding and subtracting the previous equations we can find the coefficients  $\alpha$  and  $\beta$  from the boundary conditions on  $F_s$ . It is easy to demonstrate that the ratios  $(F'_s/F_s)_{-d_s/2}$  and  $(F'_s/F_s)_{d_s/2}$  are directly related to the corresponding ratios in the ferromagnet, using the boundary conditions (5)

$$(F'_s/F_s)_{\pm d_s/2} = \frac{\gamma (F'_f/F_f)_{\pm d_s/2}}{1 \mp \xi_f \gamma_B (F'_f/F_f)_{\pm d_s/2}}. \quad (12)$$

In the next section, we will determine the critical temperature of the S-layer under general transparency conditions at the S/F interfaces. In a second part we will study the thermodynamics of the F/S/F structure at arbitrary temperature, in the limit of high and low transparencies.

#### IV. SPIN ORIENTATION DEPENDANCE OF THE CRITICAL TEMPERATURE

Close to the critical temperature and assuming that the exchange field in the ferromagnet is sufficiently strong ( $h \gg T_c$ ), the Usadel equation in the ferromagnet can be simplified as

$$-\frac{\partial^2 F_f(x, \omega)}{\partial x^2} + \frac{2ihsgn(\omega)}{D_f} F_f(x, \omega) = 0 \quad (13)$$

Using the boundary condition at a vacuum interface, we readily find the following solution for the Usadel Green functions in the parallel case (the upperscript  $P$  refers to the parallel case) for positive  $\omega$  (the case  $\omega < 0$  is obtained by making the substitution  $k_n \rightarrow k_n^*$ )

$$\begin{aligned} F_f^P(x > d_s/2) &= A \cosh k_n [x - (d_f + d_s/2)], \\ F_f^P(x < -d_s/2) &= B \cosh k_n [x + (d_f + d_s/2)], \end{aligned} \quad (14)$$

with  $k_n = (1 + i) \sqrt{\frac{h}{D_f}}$ . Analogously for the anti-parallel case (the upperscript  $A$  refers to the anti-parallel case)

$$\begin{aligned} F_f^A(x > d_s/2) &= C \cosh k_n [x - (d_f + d_s/2)], \\ F_f^A(x < -d_s/2) &= D \cosh k_n^* [x + (d_f + d_s/2)], \end{aligned} \quad (15)$$

These solutions give immediately the value of the ratios  $(F'_f/F_f)_{\pm d_s/2}$  and consequently, see (12) the ratios  $(F'_s/F_s)_{\pm d_s/2}$

$$\begin{aligned} (F'_s/F_s)_{d_s/2} &= -\frac{\gamma k_n \tanh(k_n d_f)}{1 + \xi_f k_n \gamma_B \tanh(k_n d_f)}, \\ (F'_s/F_s)_{-d_s/2}^P &= -(F'_s/F_s)_{d_s/2}, \\ (F'_s/F_s)_{-d_s/2}^A &= \frac{\gamma k_n^* \tanh(k_n^* d_f)}{1 + \xi_f k_n^* \gamma_B \tanh(k_n^* d_f)}. \end{aligned} \quad (16)$$

Then, with the help of (11), we may easily obtain the pair-breaking parameter  $\beta$ . Close to  $T_c$ , the Usadel equation may be linearized over  $F_0$  and the normal Green function is  $G_0 = \text{sgn}(\omega)$ , thus the equation (8) is simply written in first order of  $F_0$  as

$$\left[ |\omega| - \frac{D_s \beta}{2} \right] F_0 = \Delta. \quad (17)$$

Using this relation and the self-consistency equation (4) we can write down the expression for the critical temperature of the S-layer in the following general form

$$\ln \frac{T_c}{T_{c0}} = \Psi\left(\frac{1}{2}\right) - \text{Re} \Psi \left\{ \frac{1}{2} + \frac{1}{\tau(d_f) \pi T_c} \right\}, \quad (18)$$

where  $T_{c0}$  is the critical temperature of the S-layer without any proximity effect. This type of expression reminds the corresponding formula for the critical temperature of a superconductor with magnetic impurities [24], though the "magnetic scattering time"  $\tau$  may be complex in our system. It is easy to verify that in the parallel case, the effective magnetic scattering rate  $\tau^{-1}$  is indeed complex and given by the expression

$$\tau^P(d_f)^{-1} = \tau_0^{-1} \frac{(1 + i) \tanh(\widetilde{d_f})}{1 + \widetilde{\gamma_B} \tanh(\widetilde{d_f})}, \quad (19)$$

and  $\tau^{-1}$  in the anti-parallel case is real

$$\tau^A (d_f)^{-1} = \text{Re} \left( \tau^P (d_f)^{-1} \right), \quad (20)$$

here  $\widetilde{d}_f = (1 + i) \frac{d_f}{\sqrt{2}\xi_f}$ ,  $\widetilde{\gamma}_B = (1 + i) \frac{\gamma_B}{\sqrt{2}}$  and  $\tau_0^{-1} = \frac{\gamma T_{c0}}{\sqrt{2}} \left( \frac{\xi_s}{d_s} \right) \left( \frac{\xi_s}{\xi_f} \right)$ . Note that for the parallel orientation case, the critical temperature must be the same as for an S/F bilayer with S-layer thickness equals to  $d_s/2$ . The critical temperature of S/F bilayers has been recently studied in [22] and in the limit  $d_s \ll \xi_s$  the expression for  $T_c$  in [22] is indeed the same as (18) with  $\tau = \tau^P$  and  $d_s$  replaced by  $d_s/2$ . In the limit of infinite F-layers ( $d_f \rightarrow \infty$ ) and infinite transparency of the interfaces ( $\gamma_B \rightarrow 0$ ), the expression (18) reproduces the results for  $T_c$  found previously in [15,16]. If the proximity effect is weak, the parameter  $\tau_0^{-1}$  goes to zero. Expanding the Digamma function about 1/2 yields the following result in this limit

$$T_c^A = T_c^P = T_{c0} - \frac{\pi}{2} \tau^A (d_f)^{-1}. \quad (21)$$

Thus for a weak proximity effect, the shift of the transition temperature is a linear function of  $\tau_0^{-1}$  (we find here the same result as in the study of a superconducting alloy with magnetic impurities) and the difference Bautzen the critical temperatures of parallel and anti-parallel orientation appears only at the order  $\tau_0^{-3}$ .

The different kinds of obtained  $T_c(d_f)$  curves, depending on parameters of the trilayers are presented in Fig. 2 for illustration. We plot several curves for various values of  $\gamma_B$  assuming that the parameter  $\pi T_{c0} \tau_0$  is constant and equal to one. We may notice four characteristic types of  $T_c(d_f)$  behavior. The first one Fig. 2(a), at small interface transparency,  $T_c$  decays slightly non-monotonously to a finite value and the critical temperature difference between both orientations is very small. The decay presents a minimum at a particular value of  $d_f$  of the order of magnitude of  $\xi_f$ . The second one Fig. 2(b), at moderate interface transparency,  $T_c$  exhibits a reentrant behavior, it means that the superconductivity vanishes in a certain interval of  $d_f$ . For special values of the parameter  $\gamma_B$  the reentrant behavior can be observed only for the parallel orientation. The reentrance of the superconductivity

has been observed recently in Fe/V/Fe trilayers with parallel orientation of the ferromagnetic moments by Tagirov et al, see [23]. The third one Fig. 2(c), at moderately high interface transparency, the critical temperature decays monotonously and vanishes at finite value of  $d_f$ . The last type of  $T_c(d_f)$  behavior Fig. 2(d) is observed at really high interface transparency and rather thin F-layers with parallel orientation. Under these conditions the phase transition between the normal and the superconducting state presents a triple point at which the transition switch to the first order one.

In order to observe experimentally a significative spin-valve effect, it is crucial to choose the right materials and thicknesses of superconductor and F-layers to maximize  $\Delta T_c = T_c^P - T_c^A$ . Equation (18) shows that the important parameters are  $\gamma_B$ ,  $d_f$  and  $\tau_0^{-1}$ . The value of  $\tau_0^{-1}$  is directly related to the choice of the superconductor and of the ferromagnet since it is proportional to  $\gamma$  the ratio of the conductivities. This parameter does not play the crucial role in the spin-valve effect and a choice of  $\tau_0^{-1}$  around one should permit an easy observation of the effect. The choice of the thickness of the F-layer can be rather important, as shown by the curves  $T_c(d_f)$ . Indeed, due to the additional boundary condition at the interface between the ferromagnet and the vacuum, there are some interferences between incoming and reflected Cooper pairs in the F-layer. Depending on the value of  $d_f$ , these interferences can be destructive or constructive, leading to a maximum or a minimum of  $\Delta T_c$ . Finally, the curves  $T_c(d_f)$ , see Fig. 2, show that the key factor of the spin-valve effect is the transparency of the interface. For values of  $\gamma_B$  around one the effect can be easily observed whereas, if  $\gamma_B$  is an order of magnitude stronger the effect almost disappears.

In our case, both F-layers have the same thickness. The generalization to the case of F-layers of arbitrary thickness  $d_{f1}$  (for  $x < 0$ ) and  $d_{f2}$  (for  $x > 0$ ) is straightforward using (16). In the parallel case we have to make the substitution  $\tau^P(d_f)^{-1} \rightarrow \tau^P(d_{f1})^{-1} + \tau^P(d_{f2})^{-1}$  and in the anti-parallel case we have to make the substitution  $\tau^A(d_f)^{-1} \rightarrow (\tau^P(d_{f1})^{-1})^* + \tau^P(d_{f2})^{-1}$ .



## V. THERMODYNAMIC PROPERTIES OF THE STRUCTURE

In this section, we will consider the temperature dependance of the superconducting order parameter and the superconducting condensation energy in F/S/F systems. For simplicity, we concentrate on the case of F-layers of thickness  $d_f \gg \xi_f$ , which corresponds in practice to  $d_f \geq 50\text{\AA}$ . Using the classical parametrization of the Usadel equation by  $F = \sin \theta$  and  $G = \cos \theta$ , we may easily find the complex angle  $\theta(x)$  in our limit of infinite F-layers for parallel orientation [13]

$$\begin{aligned}\theta_f^P(x > d_s/2) &= 4 \arctan \left[ \tan \left( \theta_0^P/4 \right) \exp \left( -k_n (x - d_s/2) \right) \right], \\ \theta_f^P(x < -d_s/2) &= 4 \arctan \left[ \tan \left( \theta_0^P/4 \right) \exp \left( k_n (x + d_s/2) \right) \right],\end{aligned}\quad (22)$$

and for the anti-parallel one

$$\begin{aligned}\theta_f^A(x > d_s/2) &= 4 \arctan \left[ \tan \left( \theta_0^A/4 \right) \exp \left( -k_n (x - d_s/2) \right) \right], \\ \theta_f^A(x < -d_s/2) &= 4 \arctan \left[ \tan \left( \theta_0^A/4 \right) \exp \left( k_n^* (x + d_s/2) \right) \right],\end{aligned}\quad (23)$$

where  $\theta_0$  is the complex angle describing the superconducting order parameter in F-layer at S/F boundary. Note that we have assumed in the previous equations that  $\omega$  is positive (the case  $\omega < 0$  is obtained by the substitution  $k_n \rightarrow k_n^*$ ). These solutions give us immediately the ratios  $(F'_f/F_f)_{\pm d_s/2}$  and so via the boundary conditions (12), the ratios  $(F'_s/F_s)_{\pm d_s/2}$  which determines the pair-breaking parameter in the Usadel equations for the S-layer

$$\begin{aligned}(F'_s/F_s)_{d_s/2} &= -\frac{\gamma k_n \cos \theta_0}{\cos \theta_0/2 + \widetilde{\gamma}_B \cos \theta_0}, \\ (F'_s/F_s)_{-d_s/2}^P &= -(F'_s/F_s)_{d_s/2}, \\ (F'_s/F_s)_{-d_s/2}^A &= \frac{\gamma k_n^* \cos \theta_0^A}{\cos \theta_0^A/2 + \widetilde{\gamma}_B^* \cos \theta_0^A}.\end{aligned}\quad (24)$$

Using (11), we can deduce the coefficient  $\beta$  for the effective Usadel equations for both orientations ( $\omega > 0$ )

$$\beta^P = -\frac{2\gamma k_n \cos \theta_0^P}{d_s (\cos \theta_0^P/2 + \widetilde{\gamma}_B \cos \theta_0^P)},\quad (25)$$

$$\beta^A = -\left( \frac{\gamma k_n \cos \theta_0^A}{d_s (\cos \theta_0^A/2 + \widetilde{\gamma}_B \cos \theta_0^A)} + c.c. \right).\quad (26)$$

The equation (9) with (25, 26) gives, in an implicit form, the angle  $\theta$  (and so the Usadel Green) functions as a function of the Matsubara frequencies and the superconducting order parameter  $\Delta$ . Together with the self-consistency equation (4), this permits in principle to find the dependance of the superconducting order parameter on temperature and all the thermodynamics of the F/S/F system. Below, we will discuss two limiting cases which can be handled analytically: the low temperature limit and temperatures close to  $T_c$ .

### A. Low temperature behavior

When the temperature goes to zero, we may substitute the integration by a summation over Matsubara frequencies ( $\pi T \sum \rightarrow \int d\omega$ ) in the Usadel self-consistency equation for the order parameter (4)

$$\Delta = \lambda N(0) \int_{-\omega_D}^{\omega_D} F(\omega) d\omega = \lambda N(0) \int_{-\omega_D}^{\omega_D} \sin \theta d\omega, \quad (27)$$

where  $\omega_D$  of the order of magnitude of the Debye frequency is the usual cut-off in the BCS model (it will not enter in the final expressions) and  $\lambda$  is the BCS coupling constant. The integration over the Matsubara frequencies can be performed analytically when the transparency of the S/F interface is small and when it goes to infinity.

#### 1. The high transparency limit

In the high transparency limit ( $\gamma_B \rightarrow 0$ ), the angle  $\theta$ , characterizing superconductivity in the S-layer, is the same as at the S/F interface, i.e.  $\theta_0$ , see (5). Thus the Usadel equations, for parallel and anti-parallel cases become ( $\omega > 0$ )

$$\left( \omega + \frac{2(1+i)\tau_0^{-1}}{\cos \theta_0^P/2} \right) \sin \theta_0^P = \Delta \cos \theta_0^P, \quad (28)$$

$$\left( \omega + \frac{(1+i)\tau_0^{-1}}{\cos \theta_0^A/2} + c.c. \right) \sin \theta_0^A = \Delta \cos \theta_0^A. \quad (29)$$

Note that these equations are quite different from the corresponding expressions found in the case of a superconductor with magnetic impurities [24] and the analogy which worked for  $T_c$  is no longer applicable. Let us first consider the parallel case. The integral (27) can be performed analytically by changing the integration over  $\omega$  by integration over  $\theta$

$$\Delta^P = \lambda N(0) \int_{\Delta^P/\omega_D}^{\tilde{\theta}^P} \left[ \frac{\Delta^P}{\sin^2 \theta} + 2\tau_0^{-1} (1+i) \frac{\sin \theta/2}{\cos^2 \theta/2} \right] \sin \theta d\theta + c.c., \quad (30)$$

where  $\tau_0^{-1}$  is given by  $\frac{\gamma T_{c0}}{\sqrt{2}} \left( \frac{\xi_s}{d_s} \right) \left( \frac{\xi_s}{\xi_f} \right)$ , and  $\tilde{\theta}^P$  is the solution of equation

$$\Delta^P \cos \tilde{\theta}^P = 4\tau_0^{-1} (1+i) \sin (\tilde{\theta}^P/2). \quad (31)$$

In the absence of the F-layers and at zero temperature, the order parameter  $\Delta_0$  verifies (30) with  $\tau_0^{-1} = 0$ , i.e.

$$\Delta_0 = 2\lambda N(0) \int_{\Delta_0/\omega_D}^{\pi/2} \frac{\Delta_0}{\sin \theta} d\theta = 2\lambda N(0) \Delta_0 \left( -\ln \left( \tan \frac{\Delta_0}{2\omega_D} \right) \right). \quad (32)$$

Combining (30,32), we may eliminate the diverging terms when  $\theta$  goes to zero and finally, performing the remaining integration, we obtain the following explicit relation for the ratio  $\Delta^P/\Delta_0$

$$\ln \left( \frac{\Delta^P}{\Delta_0} \right) = \text{Re} \left\{ \ln \tan (\tilde{\theta}^P/2) + 4\tau_0^{-1} \frac{(1+i)}{\Delta^P} \left[ \ln \tan \left( \frac{\tilde{\theta}^P + \pi}{4} \right) - \sin (\tilde{\theta}^P/2) \right] \right\}. \quad (33)$$

Performing the same kind of calculation in the anti-parallel case we have for the ratio  $\Delta^A/\Delta_0$

$$\ln \left( \frac{\Delta^A}{\Delta_0} \right) = \ln \tan (\tilde{\theta}^A/2) + \frac{4\tau_0^{-1}}{\Delta^A} \left[ \ln \tan \left( \frac{\tilde{\theta}^A + \pi}{4} \right) - \sin (\tilde{\theta}^A/2) \right], \quad (34)$$

where  $\tilde{\theta}^A$  is the solution of

$$\Delta^A \cos \tilde{\theta}^A = 4\tau_0^{-1} \sin(\tilde{\theta}^A/2). \quad (35)$$

The density of states for one direction of spin is given by  $N_{\uparrow}(\omega) = \frac{1}{2}N(0) \text{Re}(G(\omega \rightarrow i\omega))$ , where  $N(0)$  is the total density of state in the normal state. Considering the limit  $\omega = 0$ , this relation becomes  $N_{\uparrow}(\omega) = \frac{1}{2}N(0) \text{Re}(\cos \tilde{\theta}^{A,P})$ , where  $\tilde{\theta}^P$  is given by (31) and  $\tilde{\theta}^A$  by (35). An analytical study of (31, 35) shows that the real part of the solutions  $\tilde{\theta}^{A,P}$  always exists, thus  $N_{\uparrow}(\omega)$  is finite at  $\omega = 0$ . As a result, at low temperatures and in both configurations, the superconductivity in F/S/F systems should be a gapless one.

In Fig. 3, we have plotted the order parameter in both parallel and anti-parallel case as a function of the pair-breaking parameter  $(\Delta_0\tau_0)^{-1}$ . At small exchange field or at small conductivities ratio, there is almost no difference between  $\Delta^P$  and  $\Delta^A$  and their evolution with  $(\Delta_0\tau_0)^{-1}$  is linear as in the case of superconducting alloys containing magnetic impurities, however the overall behavior in the whole temperature region is different. Naturally, the superconducting order parameter is always larger in the anti-parallel case due to the partial compensation of the exchange field effect.

The thermodynamic potential (per unit area) for both orientations can be found by integrating (33, 34)

$$\Omega^P(h, \Delta) = d_s N(0) \left( \frac{\Delta^2}{2} \ln \frac{\Delta^2}{e\Delta_0^2} - \tau_0^{-2} \text{Re}[if(X^P)] \right), \quad (36)$$

$$\Omega^A(h, \Delta) = d_s N(0) \left( \frac{\Delta^2}{2} \ln \frac{\Delta^2}{e\Delta_0^2} - \tau_0^{-2} f(X^A) \right), \quad (37)$$

where the function  $f(X)$  is defined by

$$\frac{1}{2(2(X)^2 - 1)^2} \left[ \begin{aligned} &-(X+1)^2(2X-1)^2 \ln(1+X) - (X-1)^2(2X+1)^2 \ln(1-X) \\ &+ 2X^2 \ln X + 6X^2(2X^2 - 1) \end{aligned} \right], \quad (38)$$

and while  $X^{A,P} = \sin(\tilde{\theta}^{A,P}/2)$ . Minimizing (36, 37) in respect to the order parameter at fixed exchange field gives back the self-consistency equations (33, 34) determining  $\Delta(h)$ .

Keeping in mind the fact that the free energy  $F$  of the system is equal to the thermodynamic potential when the order parameter is minimized, we have determined the difference of free energy between the parallel and the anti-parallel configuration  $F^P - F^A = \Omega^P(h, \Delta^P) - \Omega^A(h, \Delta^A)$ . The analysis of (18) in the case of infinite F-layers and high transparency of the interfaces immediately shows that the superconducting transition temperature is going to zero for

$$(\Delta_0\tau_0)^{-1} = 0.25 \text{ in the parallel case,} \quad (39)$$

$$(\Delta_0\tau_0)^{-1} = 0.175 \text{ in the anti-parallel case,} \quad (40)$$

These values naturally correspond to gaps vanishing in Fig. 3. As a result,  $F^P$  is equal to zero for  $(\Delta_0\tau_0)^{-1} > 0.25$  and  $F^A$  for  $(\Delta_0\tau_0)^{-1} > 0.175$ . In Fig. 4, we have plotted the normalized expression of  $(F^P - F^A)$ , by the free energy in the anti-parallel configuration, as a function of the parameter  $(\Delta_0\tau_0)^{-1}$ . The expression  $(F^P - F^A)$  is always positive, in conclusion, the anti-parallel configuration is always more stable than the parallel configuration.

## 2. The low transparency limit

In this limit, an expansion of equations (25, 26) with respect to  $1/\gamma_B$  can be made. In the limit of low transparency ( $\gamma_B \rightarrow \infty$ ), the order parameter in the F-layer almost completely disappears, thus the angle  $\theta_0$  describing the superconducting order parameter in F-layer at S/F boundary is small ( $\theta_0 \ll 1$ ). So, with the help of the boundary conditions (5), we find that the angle  $\theta$ , characterizing superconductivity in the S-layer, is given in this limit by  $\sin \theta = \theta_0 \widetilde{\gamma}_B / 2$ . With this relation and (11), we can easily find the expression of the coefficient  $\beta$  in both configurations

$$\beta^P = -\frac{\gamma}{\xi_f d_s \gamma_B} \left( 1 - \widetilde{\gamma}_B^{-1} + 2\widetilde{\gamma}_B^{-2} \right), \quad (41)$$

$$\beta^{AP} = (\beta^P + \beta^{P*}) / 2. \quad (42)$$

The stability of both parallel and anti-parallel configurations of the FSF trilayer in the low transparency limit can also be studied by performing the integration over  $\omega$  in the

Usadel self-consistency equation (27). We only give here the results of the corresponding calculations

$$\ln \left( \frac{\Delta^P}{\Delta_0} \right) = -\sqrt{\frac{2}{\Delta^P \tau_0 \gamma_B}} \left( 1 - \frac{1}{2\sqrt{2}\gamma_B} \right) + \frac{\pi}{4\Delta^P \tau_0 \gamma_B} \left( 1 - \frac{1}{\sqrt{2}\gamma_B} \right), \quad (43)$$

$$\ln \left( \frac{\Delta^A}{\Delta_0} \right) = -\sqrt{\frac{2}{\Delta^A \tau_0 \gamma_B}} \left( 1 - \frac{1}{2\sqrt{2}\gamma_B} - \frac{1}{16\gamma_B^2} \right) + \frac{\pi}{4\Delta^A \tau_0 \gamma_B} \left( 1 - \frac{1}{\sqrt{2}\gamma_B} \right). \quad (44)$$

Following the method presented in the previous paragraph, we obtain the expression for the thermodynamic potential

$$\Omega^{A,P}(h, \Delta) = d_s N(0) \left( \frac{\Delta^2}{2} \ln \frac{\Delta^2}{e\Delta_0^2} + a^{A,P} \Delta^{3/2} - b^{A,P} \Delta \right), \quad (45)$$

where the coefficients  $a^P = \frac{4}{3} \sqrt{\frac{2}{\tau_0 \gamma_B}} \left( 1 - \frac{1}{2\sqrt{2}\gamma_B} \right)$ ,  $a^A = \frac{4}{3} \sqrt{\frac{2}{\tau_0 \gamma_B}} \left( 1 - \frac{1}{2\sqrt{2}\gamma_B} - \frac{1}{16\gamma_B^2} \right)$  and  $b^{A,P} = \frac{\pi}{2\tau_0 \gamma_B} \left( 1 - \frac{1}{\sqrt{2}\gamma_B} \right)$ . The term containing  $\gamma_B^{-2}$  in  $a^A$  contributes to the stabilization of the anti-parallel configuration compared to the parallel configuration. Although, as it follows from (43, 44, 45), the orientation dependent relative variation of the order parameter and condensation energy is very small

$$\frac{\Delta^P - \Delta^A}{\Delta_0} \sim \frac{F^P - F^A}{F_0} \sim \frac{\gamma_B^{-5/2}}{\sqrt{\Delta_0 \tau_0}}. \quad (46)$$

## B. Free energy, entropy and specific heat of the trilayer close to $T_c$

At the transition temperature, the order parameter  $\Delta$  goes to zero and the Green functions  $F$  and  $G$  go respectively to 0 and  $\text{sgn}(\omega)$ . In the limit of high S/F interfaces transparency, we may use (28, 29) and develop all the quantities around  $T_c$  to obtain an expansion of  $F$  in powers of  $\Delta$

$$F = \frac{\Delta}{|\omega| + \epsilon(\omega)} - \frac{\Delta^3}{2(|\omega| + \epsilon(\omega))^3} - \frac{\Delta^3 \epsilon(\omega)}{8(|\omega| + \epsilon(\omega))^4} + o(\Delta^5), \quad (47)$$

where for the parallel case  $\epsilon = \epsilon^P(\omega) = \frac{2(1+i)\tau_0^{-1}}{\cos \theta_P/2}$  while in the parallel case  $\epsilon = \epsilon^A(\omega) = \frac{\epsilon^P(\omega) + \epsilon^P(\omega)^*}{2}$ . Thus, using expression (47) and the self-consistency equation, we may directly

obtain the dependance of the order parameter with the temperature. In the anti-parallel case we have

$$-\ln\left(\frac{T}{T_{c0}}\right) = \Psi\left(\frac{1}{2} + \frac{1}{\pi\tau_0 T}\right) - \Psi\left(\frac{1}{2}\right) + \left(\frac{\Delta}{2\pi T}\right)^2 g_1\left(\frac{1}{\pi\tau_0 T}\right), \quad (48)$$

and in the parallel case

$$-\ln\left(\frac{T}{T_{c0}}\right) = \text{Re}\Psi\left(\frac{1}{2} + \frac{(1+i)}{\pi\tau_0 T}\right) - \Psi\left(\frac{1}{2}\right) + \left(\frac{\Delta}{2\pi T}\right)^2 \text{Re}g_1\left[\frac{(1+i)}{\pi\tau_0 T}\right], \quad (49)$$

where the function  $g_1(x) = -\frac{1}{4}\Psi^{(2)}\left(\frac{1}{2} + x\right) + \frac{x}{48}\Psi^{(3)}\left(\frac{1}{2} + x\right)$ . It's important to note that the function  $g_1(x)$  is positive for all values of  $(\pi\tau_0 T)^{-1}$ , so the superconducting phase transition is always a second order one for  $d_f \gg \xi_f$ . The transition temperature of the superconductor in contact with the F-layers is determined by putting  $\Delta = 0$  in the previous equations and gives back the results of the previous section and of [15,16], in the limit of large F-layer and large transparency of the interfaces. Simplifying (48, 49) using (18), and defining the function  $g_2(x) = 1 - x\Psi^{(1)}\left(\frac{1}{2} + x\right)$ , we get

$$\Delta_A^2 = (2\pi T_c^A)^2 \frac{g_2\left[(\pi\tau_0 T_c^A)^{-1}\right]}{g_1\left[(\pi\tau_0 T_c^A)^{-1}\right]} \left(1 - \frac{T}{T_c^A}\right), \quad (50)$$

$$\Delta_P^2 = (2\pi T_c^P)^2 \frac{\text{Re}g_2\left[(\pi\tau_0 T_c^P)^{-1}\right]}{\text{Re}g_1\left[(\pi\tau_0 T_c^P)^{-1}\right]} \left(1 - \frac{T}{T_c^P}\right). \quad (51)$$

This shows that the order parameters increases as  $(1 - T/T_c)^{1/2}$  when the temperature is sufficiently low. The free energy of the system is simply given by, see [21],

$$F_s - F_n = \Delta F = - \int_0^\lambda \frac{\Delta^2}{\lambda_1^2} d\lambda_1 = \int_0^\Delta \Delta_1^2 \frac{d\left(\frac{1}{\lambda}\right)}{d\Delta_1} d\Delta_1. \quad (52)$$

Using the relation  $\delta(1/\lambda) = -N(0)\delta T_{c0}/T_{c0}$ , see [21], and equations (50, 51), we obtain  $\frac{d(\lambda^{-1})}{d\Delta} = -\frac{\Delta N(0)}{2\pi^2 T_c^2} g_1\left[(\pi\tau_0 T_c)^{-1}\right]$ . Thus the calculation of the free energy is straightforward

$$\Delta F^A = -2N(0)\pi^2 (T_c^A)^2 \frac{g_2^2 \left[ (\pi\tau_0 T_c^A)^{-1} \right]}{g_1 \left[ (\pi\tau_0 T_c^A)^{-1} \right]} \left( 1 - \frac{T}{T_c^A} \right)^2, \quad (53)$$

$$\Delta F^P = -2N(0)\pi^2 (T_c^P)^2 \frac{\left\{ \text{Re } g_2 \left[ (\pi\tau_0 T_c^P)^{-1} \right] \right\}^2}{\text{Re } g_1 \left[ (\pi\tau_0 T_c^P)^{-1} \right]} \left( 1 - \frac{T}{T_c^P} \right)^2. \quad (54)$$

From the above equations, the entropy and the heat capacity are obtained using  $S = - \left( \frac{\partial F}{\partial T} \right)_h$  and  $C = -T \left( \frac{\partial^2 F}{\partial T^2} \right)_h$ . We present the results obtained for the heat capacity only

$$\Delta C^P (T_c^P) = 4\pi^2 N(0) T_c^P \frac{\left\{ \text{Re } g_2 \left[ (\pi\tau_0 T_c^P)^{-1} \right] \right\}^2}{\text{Re } g_1 \left[ (\pi\tau_0 T_c^P)^{-1} \right]}, \quad (55)$$

$$\Delta C^A (T_c^A) = 4\pi^2 N(0) T_c^A \frac{g_2^2 \left[ (\pi\tau_0 T_c^A)^{-1} \right]}{g_1 \left[ (\pi\tau_0 T_c^A)^{-1} \right]}. \quad (56)$$

The jump of the specific heat at the transition decreases monotonically as  $T_c$  decreases (i.e. as the pair-breaking effect of the F-layers increases). The corresponding results are plotted in Fig. 5, where the jump of the specific heat at  $T_c$  is normalized by the jump of the specific heat at  $T_{c0}$  the critical temperature without any proximity effect.

## VI. CONCLUSION

We have considered the properties of F/S/F spin-valve systems and presented their general theoretical description for the most interesting case of thin superconducting layer. The spin-valve effect occurs to be very strongly dependent on the S/F interface transparency. So, to observe it on experiment it is necessary to chose superconductor-ferromagnet systems with a low barrier at the interface. The oscillatory-like  $T_c$  dependance on the F-layer thickness  $d_f$  gives the optimum condition of spin-valve observation for  $d_f \sim \xi_f$  i.e.  $10 - 50 \text{ \AA}$ , but the situation remains qualitatively the same for higher thickness too. The maximum gain in the superconducting energy corresponds to the anti-parallel configuration, and this gain may be of the same order of magnitude as the superconducting condensation energy itself. So we may except that without external applied field, parallel configuration will be



unstable. Therefore, with the decrease of the temperature below  $T_c$ , the transition from parallel to anti-parallel configuration may be observed. Since it would depend on the magnetic coercitivity force, thin F-layers would be "a priori" more suitable to observe such effect. A very interesting situation can be also observed when the Curie temperature is lower than the superconducting critical temperature. In such a case we may expect the spontaneous appearance of the anti-parallel configuration by decreasing the temperature. It is worth to note that in the case when the domain wall energy is small, the formation of short length-scale magnetic domains could occur at the contact of the ferromagnet and the superconductor [25,26].

In conclusion, the F/S/F trilayer systems reveal strong interferences between superconducting and magnetic effects. They could be quite interesting for application as a very small magnetic field may strongly influence the superconducting characteristics via the spin-valve effect.

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FIG. 1. Geometry of the F/S/F sandwich. The thickness of the S-layer is  $d_s$  and  $d_f$  is the thickness of the F-layers.

FIG. 2. Characteristic types of  $T_c(d_f)$  behavior. The thickness of the F-layer is normalized to the F-layer characteristic length  $\xi_f$ . The parameter  $\pi\tau_0 T_{c0}$  is chosen constant and equal to one. The full line corresponds to the anti-parallel case, the dotted line to the parallel case. One can distinguish four characteristic types of  $T_c(d_f)$  behavior: (a) and (b)

monotonic decay to  $T_c = 0$  with (a) or without (b) switching to a first order transition in the parallel case, (c) reentrant behavior for the parallel orientation, (d) nonmonotonic decay to a finite value of  $T_c$ .

FIG. 3. The order parameter  $\Delta$  normalized by its value in absence of proximity effect  $\Delta_0$  in both parallel and anti-parallel case as a function of the pair breaking parameter  $(\tau_0\Delta_0)^{-1}$ .

FIG. 4. Normalized value of the difference of free energy between the parallel and the anti-parallel configurations plotted as a function of the parameter  $(\tau_0\Delta_0)^{-1}$ . For  $(\tau_0\Delta_0)^{-1} \geq 0.175$ , the superconducting transition temperature is equal to zero in the parallel configuration.

FIG. 5. Discontinuity of the specific heat at the critical temperature versus  $T_c/T_{c0}$ . The full line corresponds to the anti-parallel case, the dotted line to the parallel case.











